UNCLASSIFIED

Defense Technical Information Center
Compilation Part Notice

ADP013062

TITLE: Thin Film Photonic Crystals

DISTRIBUTION: Approved for public release, distribution unlimited
Availability: Hard copy only.

This paper is part of the following report:

TITLE: Nanostructures: Physics and Technology International Symposium [8th] Held in St. Petersburg, Russia on June 19-23, 2000 Proceedings

To order the complete compilation report, use: ADA407315

The component part is provided here to allow users access to individually authored sections of proceedings, annals, symposia, etc. However, the component should be considered within the context of the overall compilation report and not as a stand-alone technical report.

The following component part numbers comprise the compilation report:
ADP013002 thru ADP013146

UNCLASSIFIED

8th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 19–23, 2000
© 2000 Ioffe Institute

Thin film photonic crystals

Clivia M. Sotomayor Torres†, Torsten Maka†, Manfred Müller‡,
Rudolf Zentel‡ and Sergei G. Romanov†§
† Institute of Materials Science and Dept. of Electrical Engineering,
University of Wuppertal, Gauss-str. 20, 42097 Wuppertal, Germany
‡ Institute of Materials Science and Dept. Chemistry, University of Wuppertal,
Gauss-str. 20, 42097 Wuppertal, Germany
§ Ioffe Physico-Technical Institute, St Petersburg, Russia

Introduction

The fabrication of 3-dimensional photonic bandgap (PBG) based light moulding and emitting devices requires (i) highly crystalline periodical structures, (ii) emitters incorporated in a PBG environment and (iii) emission band tuned to the optical stop-band of the photonic crystal. The combination of the physically separated (a) light source with a high emission yield and an emission band narrower than the PBG bandwidth and (b) photonic crystal with the refractive index contrast (RIC) large enough to open an omnidirectional bandgap is possible to approach using 3D photonic templates, partucularly, opals infilled with a light emitting material [1]. We have extended this approach by preparing opal-like structures from polymers. Due to the low RI and the incomplete PBG in polymer-made structures, polymeric photonic crystals possess strongly anisotropic optical properties. From the other hand, crystalline quality packages of polymeric balls can be processed further to prepare semiconductor replicas of a higher refractive index contrast (RIC) and the optimised filling factor. Organic dyes embedded in polymers are known as effective luminescent materials [2–4]. Moreover, they offer excellent flexibility to match the stop-band with the emission band of the dye without raising dramatically the imaginary part of the dielectric function. We discuss here the optical properties of several polymeric photonic materials and their replicas. Taken the anisotropy into account, angular resolved spectroscopy [5–6] has been used to find the PBG effect in a particular direction of the photonic crystal and to study the change of the spontaneous emission.

1. Opal-like thin film crystals

PMMA balls for polymer photonic crystals were prepared by a modified emulsion polymerisation from the monomer-solution. In some cases a small amount of fluorescent dye (Coumarin 6) has been added to the solution to study the PBG effect upon emission from photonic crystal. The ball diameter was controlled by varying the polymerisation time. Particles of larger size were separated by filtration and centrifugation. Films of several cm^2 area were deposited on hydrophilized microscope slides. Balls are self-assembled in the face-centered cubic fcc package with (111) plane along the substrate. These thin film polymeric crystals consist of ca. 30–50 monolayers and possess domains extending over hundreds of micrometers. Dye molecules are distributed homogeneously in the balls and their concentration can be varied in a controlled manner. The resulting crystal offers the advantage of containing light emitting molecules inside the photonic structure while preserving the RIC of the host polymer. In Fig. 1 an SEM micrograph of the initial photonic structure is shown. The monodispersity of the spheres is less than 10% but shows smaller

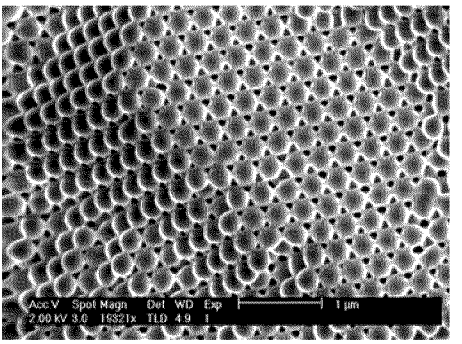

Fig. 1. SEM micrograph showing a film of fcc packed PMMA balls on a glass substrate.

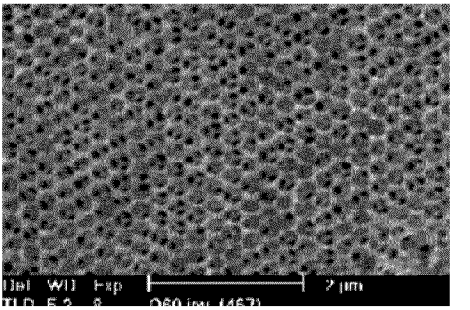

Fig. 2. SEM micrograph of inverted opal structure.

variations on short length scales. The vapour phase chemical deposition was applied to fill the polymeric opal-like film with SnS_2. Polymeric balls were dissolved after completing the impregnation thus leaving the semiconductor replica film on the glass substrate (Fig. 2). The filling factor of the PMMA-air structure $f_{\text{ball}} = 0.74$ is far from being optimal and the RIC is about 1.5 to 1. In the inverse structure the fraction of high RI material $1 - f_{\text{ball}}$ is closer to the optimum value, whereas the RIC depends on the actual density of a semiconductor and varies from 1.4–1.9 to 1.

2. Diffraction

The transmission and reflectance spectra were measured by illuminating the sample with white light. The angular dispersion of the Bragg diffracted light was studied by measuring reflectance spectra at different angles θ between the (111) axis of fcc crystal and the beam axis by collecting the scattered light within a solid angle of 2°. Changes in the light diffraction have been quantified using the Bragg law $\lambda_{(111)} = 2n_{\text{eff}} \cdot 0.81D$, with n_{eff} being the effective RI of the composite and D the diameter of opal balls. The n_{eff} was determined using the effective medium approximation $n_{\text{eff}} = n_1 f_{\text{ball}} + n_2(1 - f_{\text{ball}})$, n_i is RIs of crystal components [7, 8]. It is known, that the angular dispersion of diffraction peaks mimics the dispersion of the stop-band in the $E - k$ space [9]. Ball diameters extracted from the Bragg diffraction are in good agreement with SEM data, if the RI for PMMA $n = 1.4893$ applies [10].

Both changes of the effective RI and the filling factor of the high-RI material (f_{hRI}) lead to a shift of the Bragg resonance and the increase of the width of the photonic bandgap in semiconductor replica as compared with polymeric template. A comparison of angle-resolved reflectance spectra is shown in Fig. 3. The reflectance of the polymeric opal shows a relative stop-bandwidth of $\Delta E/E_0 \approx 5\%$. The "blue" shift of the Bragg resonance in the replica due to the decrease of the filling factor is balanced by the "red" shift due to the increase of the RIC. Simulateneously, both factors serve to increase the photonic bandwidth to $\Delta\lambda/\lambda_0 = 13\%$. It is worth mentioning that the distortion of the lattice, which is bigger in replica, also contributes to the broadening of the Bragg peaks.

The rate of the angular dispersion of the stop-band is governed by the effective RI. Correspondingly, it is stronger for replica, but decreases with the increase of the density of SnS_2. However, the total improvement of the PBG in the replica as compared with the polymeric template is the result of the much wider stop-band, that allows to overlap them

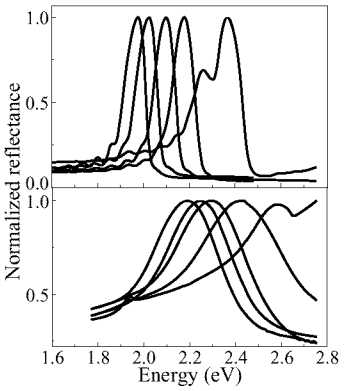

Fig. 3. Angle resolved reflectance spectra of PMMA opal film (top) and inverted SnS_2 opal film (bottom). Spectra for angles 5, 20, 30, 40 and 50 degrees from left to right.

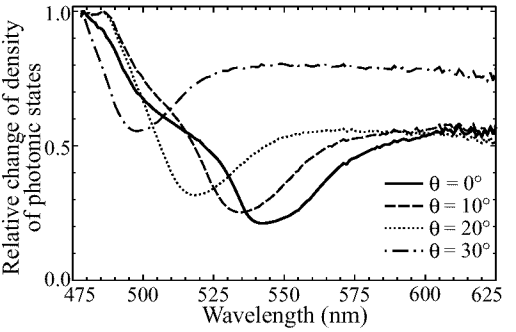

Fig. 4. Relative dip in density of photonic states obtained from PL measurements.

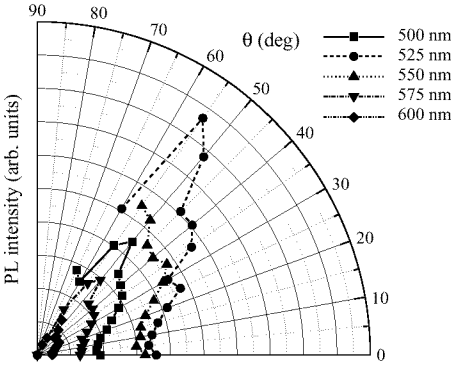

Fig. 5. Directionality diagram of the emission from polymer-dye reference sample.

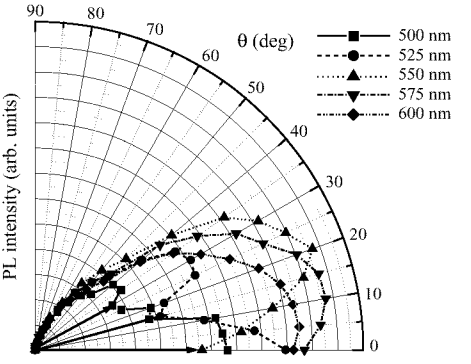

Fig. 6. Directionality diagram of the emission from dye-polymer-opal film.

more effectively for different angles. Expressing this overlap in terms of the ratio of the stop-bandwidth ΔE to the shift of the resonance frequency E_{shift} as $\delta = \Delta E / E_{\text{shift}}$, the δ is factor improves from 0.5 to 1.3.

3. Photoluminescence

Bragg configuration was used to study the photoluminescence (PL) as the angle was varied between $\theta = 0^\circ$ and $\theta = 50^\circ$ the PL was collected within a 6° solid angle. PBG reduces the probability of the radiative recombination within the stop-band and, consequently, the PL spectrum shows the dip, which spectral position depends on the angle of the emission detection. To demonstrate the relative changes in the density of photonic states in the PBG region PL spectra are shown normalised to the PL spectrum on unstructured Coumarin-PMMA film (Fig. 4).

The anisotropy of the photonic band structure leads to the self-focusing of the emission (Fig. 5). The fingerprints of the PBG are reproduced at different angles for different

frequencies as a dip in the directionality diagram. In contrast the similar diagram of the dye-polymer film shows no obvious wavelength dependence (Fig. 6). The similar effect was mention previously for dye-polymer loaded opals [11]. By analogy with the superprism effect [12], the emission self-focusing in the incomplete photonic crystal is related to the topology of dispersion planes for electromagnetic waves with the frequency near the stop-band, because the vector of the group velocity of propagating wave is always normal to the dispersion plane. Accordingly, the focusing rate depends on the energy.

4. Conclusions

Two photonic crystals have been prepared based on polymer opal-like films. Both polymer and semiconductor structures demonstrate similar photonic behaviour. The width and dispersion of the stop-band has been greatly improved by decreasing the the filling factor of a high RI component and increasing the RIC. An antenna like effect — the focusing of emission from incomplete photonic crystals — has been observed and explained as the result of specific dispersion of the electromagnetic waves in the frequency range of PBG.

References

[1] S. G. Romanov and C. M. Sotomayor Torres, in *Handbook of Nanostructured Materials and Technology*, ed H. S. Nalwa, vol 4, ch 4, pp 231–323, Acad. Press, 1999.

[2] K. Yoshino, K. Tada, M. Ozaki, A. A. Zakhidov and R. H. Baughman, *Jpn. J .Appl. Phys.* **36**, L714, (1997);
K. Yoshino, S. B. Lee, S. Tatsuhara, Y. Kawagishi, M. Ozaki and A. A. Zakhidov, *Appl. Phys. Lett.* **73**, 3506, (1998).

[3] T. Yamasaki and T. Tsutsui, *Appl. Phys. Lett.* **72**, 1957, (1998).

[4] E. P. Petrov, V. N. Bogomolov, I. I. Kalosha and S. V. Gaponenko, *Phys. Rev. Lett.* **81**, 77, (1998).

[5] S. G. Romanov, *J. Nonlinear Opt. Phys. Mater.* **7**, 181, (1998).

[6] S. G. Romanov, A. V. Fokin and R. M. De La Rue, *Appl. Phys. Lett.* **74**, 1821, (1999).

[7] W. L. Vos, R. Sprik, A. von Blaaderen, A. Imhof, A. Lagendijk and G. H. Wegdam, *Phys. Rev. B* **53**, 16231 (1996);
R. D. Pradhan, I. I. Tarhan and G. H. Watson, *Phys. Rev. B* **54**, 13721, (1996).

[8] C. Lopez, H. Miguez, L. Vazquez, F. Meseguer, R. Mayoral and M. Ocana, *Superlat. Microstruct.* **22**, 399 (1997).

[9] V. Yannopapas, N. Stefanou and A. Modinos, *J. Phys.: Cond. Matter.* **9**, 10261, (1998).

[10] J. Braderup and E.H. Immergut, *Polymer Handbook* (3d Ed), New York, p VI/459, 1989.

[11] S. G. Romanov, T. Maka, C. M. Sotomayor Torres, M. Mueller, D. Allard and R. Zentel, *Appl. Phys. Lett.* **75**, 1057 (1999).

[12] H. Kosaka, T. Kawashimaa, A. Tomita, M. Notomi, T. Tamamura, T. Sato and S. Kawakami, *Phys. Rev. B* **58**, R10096, (1998).